Light and electric field control of ferromagnetism in magnetic quantum structures

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A strong influence of illumination and electric bias on the Curie temperature and saturation value of the magnetization is demonstrated for semiconductor structures containing a modulation-doped p-type $Cd_{0.96}Mn_{0.04}Te$ quantum well placed in various built-in electric fields. It is shown that both light beam and bias voltage generate an isothermal and reversible cross-over between the paramagnetic and ferromagnetic phases, in the way that is predetermined by the structure design. The observed behavior is in quantitative agreement with the expectations for systems, in which ferromagnetic interactions are mediated by the weakly disordered two-dimensional hole liquid.

Soon after the discovery of carrier-controlled ferromagnetism in Mn-doped III-V [1] and II-VI [2] semiconductor compounds, it has become clear that these systems offer unprecedented opportunities to exploit the powerful methods developed for tuning carrier densities in semiconductor quantum structures, in order to control the magnetic characteristics in these systems [2, 3, 4, 5, 6]. Such a control opens new prospects for information storage and processing, as well as it makes it possible to examine the behavior of strongly correlated systems as a function of externally controllable parameters. In the case of III-V magnetic semiconductors, Koshihara *et al.* [3] detected an enhancement of ferromagnetism by illumination of an (In,Mn)As/GaSb heterostructure, an effect assigned to the presence of an interfacial electric field that drives the photo-holes to the magnetically active (In,Mn)As layer. More recently, Ohno *et al.* [6] demonstrated that a gate voltage of ± 125 V changes the Curie temperature T_C by about 1 K in a field-effect transistor structure containing an (In,Mn)As quantum well (QW).

In the case of II-VI diluted magnetic semiconductors (DMS), Mn does not introduce any carriers. Hence, hole-mediated ferromagnetic interactions can be induced by modulation-doping of heterostructures [7]. Due to the valence band structure, T_C is typically lower in II-VI than in III-V DMS. At the same time, however, it may be expected [2, 5] that, owing to the small background hole density, the strength of the carrier mediated ferromagnetic interactions can be tuned over a wider range in II-VI than in III-V DMS.

In this paper, we present photoluminescence (PL) studies of modulation-doped p-type (Cd,Mn)Te QW. The (Cd,Zn,Mg)Te barriers are doped either p- or n-type, so that p-i-p or p-i-n structures are formed. The QW in these systems are ferromagnetic below about 3 K. We show that, depending of the sample layout, the ferromagnetism is either destroyed or enhanced during illumination by photons with energy greater than the band gap of the barrier material. In both cases, the switching process is isothermal and reversible. Moreover, we demonstrate that the reverse biasing of the p-i-n diode by a voltage smaller than 1 V turns the ferromagnet into a paramagnetic material. Importantly, this strong effect of light and electric field can be readily explained by considering the distribution of carriers and photo-carriers in the p-i-p and p-i-n structures. At the same time, by tracing how the system properties vary with the hole density in the QW, we identify processes accounting for the magnitude of the Curie temperature and spontaneous magnetization in this low-dimensional ferromagnetic system.

Our samples were grown coherently onto a $Cd_{0.88}Zn_{0.12}Te$ substrate by molecular beam epitaxy, exploiting previous expertise in p-type [8] and n-type [9] modulation doping of tellurides. As shown in Fig. 1, the structures contain a single 8 nm QW of (Cd,Mn)Te, with typically 3% to 5% Mn and (Cd,Mg,Zn)Te barriers. In the back barrier of the p-i-p structure, a nitrogen-doped p-type layer is inserted at 100 nm from the QW. A hole gas in the QW is created by doping the top barrier with nitrogen acceptors of concentration exceeding 10^{17} cm^{-3}. The corresponding spacer layer thickness is 20 nm. In the p-i-n diode, the back barrier doped with aluminum (n-type) resides 320 nm away from the QW, and the spacer between the QW and the p-doped layer is diminished to 10 nm. This leads to a hole density in the QW of about 2×10^{11} cm^{-2} in both p-i-p and p-i-n structures, so that the carriers occupy only the ground-state heavy-hole subband. A semi-transparent gold film is evaporated on top of the p-i-n diodes, and then 2×2 mm^2 squares are formed by Ar-ion etching down to the n-type layer, a procedure followed by the deposition of In contacts. In these diodes, non-linear current-voltage characteristics are observed up to room temperature.

Following the established procedure [2, 5, 10, 11], we probe the properties of the system by PL and its excitation (PLE). Owing to the exchange coupling between the band carriers and the Mn spins, the PL line splitting is

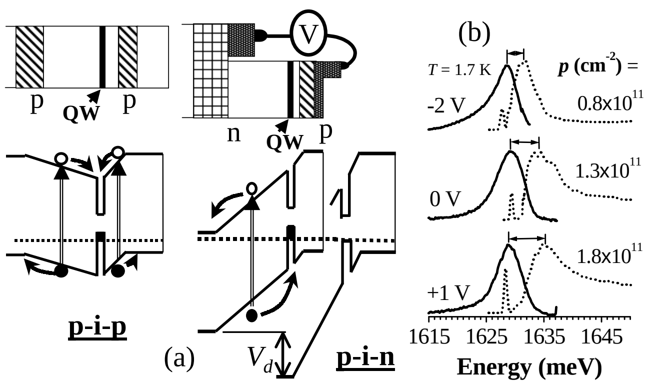

FIG. 1: (a)Layout of p-i-p and p-i-n structures containing a (Cd,Mn)Te quantum well, and band alignments with and without bias voltage V_d, showing the expected migration of the photocarriers. (b) PL (solid line) and PLE (dashed line) showing the Moss-Burstein shift (arrows), *versus* bias voltage V_d, on a p-i-n structure containing a $Cd_{0.995}Mn_{0.005}Te$ QW. The low energy feature in PLE is the laser peak.

proportional to the Mn magnetization.This allows us to measure the magnetization locally, in a single QW, with a much higher sensitivity than using a conventional magnetometer. PL is excited by an Al_2O_3:Ti or HeNe laser, whose photon energy is below the barrier gap, and the output power is kept below 9 mW/cm^2. An additional above-barrier illumination, which aims at affecting the ferromagnetism (by increasing or reducing the hole density, as explained below), is provided either by a halogen lamp screened by one blue and a variable number of gray filters, or by an Ar-ion laser. The Moss-Burstein shift between the PL and PLE lines serves us to determine the hole density [11] at given values of illumination intensity and bias voltage. To increase the accuracy, these measurements were also carried out in a magnetic field over 1 T, in which the hole gas is entirely spin polarized, so that the magnitude of the shift is doubled. Fig.1b shows the influence of the bias voltage V_d, on the Moss-Burstein shift (arrows) for a sample with only 0.5% Mn in the QW so that no ferromagnetic ordering is observed. This procedure appears to provide an accurate evaluation of the relative hole density, whereas its absolute value is determined within a factor of 2. It was checked that for the depleted QW both spectral position and giant Zeeman splitting of the exciton reflectivity line are consistent with those expected for the nominal values of QW width and Mn concentration (3% to 5%). Furthermore, an increase of the spin temperature [12] can be deduced from the Zeeman splitting but only for much higher illumination intensities than in the present study.

Figure 2 presents PL spectra collected for p-i-p and p-i-n structures in the absence of an external magnetic field at various temperatures, illumination intensities, and bias voltages. The PL line corresponds to the e1→ hh1 transitions. Its splitting, accompanied by a red shift of its lower component [2, 5], signals the transition to an ordered phase. The splitting energy is proportional to the spontaneous magnetization within a magnetic domain. Since the easy axis is oriented along the growth direction, the emitted light contains two circularly polarized components of equal intensity corresponding to the two possible orientations of the magnetic domains. Applying a small field in the Faraday geometry results in variations of the splitting and circular polarization of the lines in agreement with the evolution expected from such domains [5]. As shown, the phase transition occurs not only on lowering the temperature [Figs. 2(a) and 2(c)], but also isothermally, on changing the illumination [Fig. 2(b)] or bias voltage [from Fig. 2(c) to Fig.2 (d)]. Remarkably, while above barrier illumination destroys ferromagnetism in the p-i-p structures [Fig. 2(b)], it enhances the spontaneous magnetization in the p-i-n diodes [dotted line in Fig. 2(c)]. Since in the systems in question ferromagnetic interactions are mediated by holes, we assign the observed behavior to the influence of illumination and bias voltage on the hole density in the QW which contains the Mn spins. To put these considerations onto a more quantitative basis we will first relate the illumination intensity and bias voltage to the hole density, and then examine how the latter determines T_{C} and spontaneous magnetization.

As shown for similar p-i-p structures with a lower Mn content [11], photons of energy larger than the barrier bandgap reduce the hole density in the QW. According to Fig. 1, the photo-electrons created in this way migrate to the QW, where they recombine with the pre-existing holes. At the same time, photo-holes are trapped in the barrier layers, and tunnel rather slowly back to the QW. The interplay of these processes determines the steady hole density. In agreement with the previous studies [11], the relation between the light intensity I and the hole density p determined from the Moss-Burstein shift is well described by the formula expected for tunneling through the triangular barrier arising due to the electric field induced by the presence of the 2D hole gas in the QW, $I = A(p - p_0)\exp(-\beta\sqrt{p})$, where p_0 is the hole density without illumination, and the parameters A and β are proportional to the rates of electron migration and hole tunneling, respectively. We find their values to be similar to those determined previously [11], *e.g.*, $\beta = 3.34 \times 10^7$ m. Moreover, photo-carriers created directly by PL excitation into the QW , recombine fast so that they do not affect the carrier density [11].

Turning to the p-i-n diodes, the band structure (Fig. 1), and the fact that the penetration length of light is larger than one micrometer, show that the built-in electric field drives the majority of the photo-holes towards the QW. This causes an enhancement of ferromagnetism under illumination, an effect visible in Fig. 2(c). In addition, biasing the p-i-n diodes controls the QW hole density: the density, evaluated from the Moss-Burstein shift,

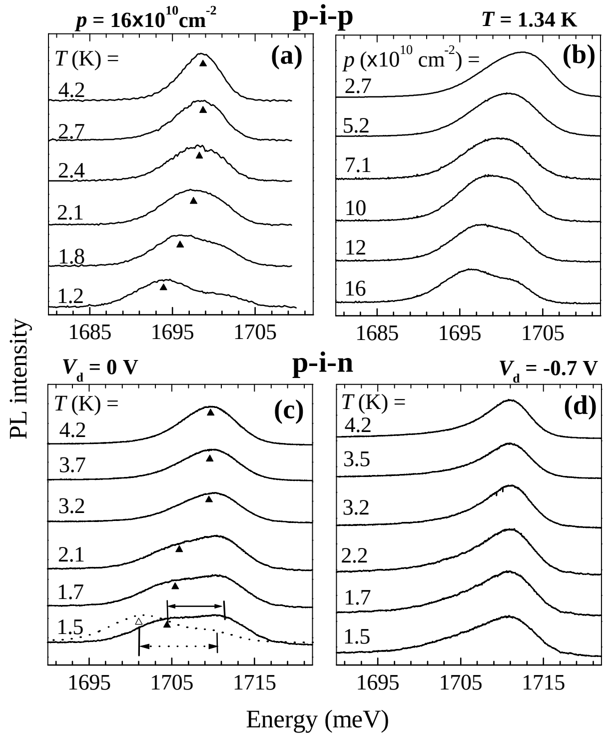

FIG. 2: PL spectra for a modulation-doped p-type $Cd_{0.96}Mn_{0.04}Te$ QW located in a p-i-p structure and a modulation-doped p-type $Cd_{0.95}Mn_{0.05}Te$ QW in a p-i-n diode; (a) p-i-p structure without additional illumination (i.e., constant hole density) at various temperatures; (b) same p-i-p structure with above barrier illumination (to reduce the hole density) at fixed temperature; (c) p-i-n structure without bias at various temperatures: the hole density is constant (solid lines) or increased (dotted line) by additional Ar-ion laser illumination; (d) p-i-n structure with a -0.7 V bias (depleted QW) at various temperatures. Splitting and shift of the lines mark the transition to the ferromagnetic phase.

changes from $p_0 \approx 2 \times 10^{11}$ cm^{-2} at zero bias down to zero at -0.7 V. Accordingly, at this bias, no signature of ferromagnetism is observed in the PL spectra down to 1.5 K, as shown in Fig. 2(d). Knowing the sample layout, we can estimate the bias V_d, which has to be applied in order to deplete the QW entirely. The main contribution is that of the electric field across the capacitance formed by the QW and the n-type layer. For the dielectric constant of CdTe, $\epsilon = 10$, this leads to $V_d \approx e p_0 L/\epsilon_o \epsilon = 1$ V which reproduces well the experimental value.

An important aspect of our data is that they provide detailed information on the dependence of T_C and the spontaneous magnetization on the hole density p. There are two ways of crossing the phase boundary in our system: by varying the temperature T at constant p, or by varying p at constant T. The experimental values of T_C have been obtained by tracing the position of the lower PL line as a function of T at constant p, as shown in Fig. 3(a). This procedure avoids uncertainties associated with the renormalization of the PL energy by carrier-carrier correlation, which depends more on p than on T, an effect visible in Fig. 2(b). A mean-field model for T_C in low-dimensional structures has been derived by some of the present authors [2, 7] and others [13, 14]. The applicability of the mean-field approximation (MFA) is justified by the long-range character of the ferromagnetic interactions in question. In the theory adopted here [2, 7] the contribution of short range antiferromagnetic interactions is included in terms of an effective spin density and with an effective Curie-Weiss temperature which is negative (noted $-T_\mathrm{AF}$ hereafter) as measured for undoped DMS. As a result, the Curie-Weiss temperature is given by $T_\mathrm{C} = T_\mathrm{F} - T_\mathrm{AF}$, where T_F is proportional to the Pauli susceptibility of the hole liquid $\tilde{\chi}_h$. Hence in a degenerate gas, $\tilde{\chi}_h$ is proportional to the density of states at Fermi energy, which is independent of p for an ideal 2D gas [7]. The hole-hole interactions are incorporated into $\tilde{\chi}_h$ according to the Fermi liquid theory. In the following, when evaluating $\tilde{\chi}_h$ at such low values of the hole density [5], the assumption that the hole liquid is in the highly degenerate limit is relaxed, and the electrostatic disorder is taken into account by a gaussian broadening of the hole density of states. Then, T_C evolves smoothly from $-T_\mathrm{AF}$ to the constant value pertaining to the clean degenerate 2D liquid.

As shown in Fig. 3(b), there is a good agreement between experimental and theoretical values of T_C as a function of the hole density. The theoretical curve is drawn with two adjustable parameters: the Fermi liquid parameter $A_F = 2.1$ and the full-width at half maximum of the density-of-states gaussian broadening $\Gamma = 1.7$ meV. Such a two-fold enhancement of the Pauli susceptibility by the interactions, as implied by the magnitude of A_F, is to be expected for the hole density range in question [14]. According to Fig. 3(b), it is principally the non-zero value of Γ which accounts for the lowering of T_C observed in the regime of small hole densities. Indeed, the value of Γ determined here compares favorably with the activation energy of 1.5 meV, determined for positively charged excitons in a similar CdTe QW [15]: these charged excitons were thought to be trapped by potential fluctuations brought about by acceptors residing at 50 nm from the QW on its both sides.

It has been shown [7] that, in the ideal 2D case, the hole gas should be completely polarized immediately below T_C. Therefore, the magnetization of the Mn spins induced by the molecular field of the spin polarized holes is expected to increase linearly with the hole density. Experimental values of the zero-field splitting at 1.35 K, plotted versus the hole density in Fig. 3(c), corroborate this expectation in the density range where the zero-field splitting is observed. The small difference between the experimental values and the results of the mean-field model [7] may suggest a possible influence of carrier-carrier correlation on optical spectra. However,

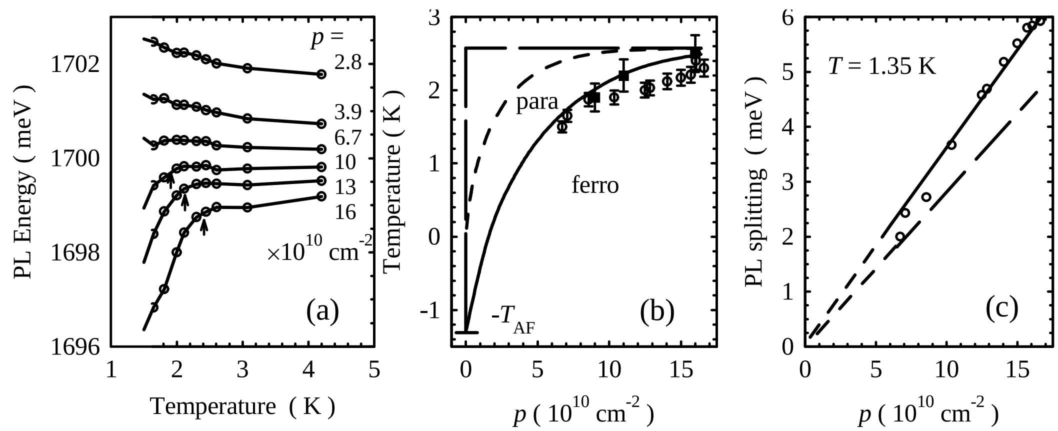

FIG. 3: (a) Temperature dependence of the low-energy peak of the PL spectra for selected values of the hole density (changed by illumination) for a p-type $Cd_{0.96}Mn_{0.04}Te$ QW. Arrows indicate the points taken as indicating the critical temperature of the ferromagnetic transition (T_C). (b) T_C *versus* hole density for two samples (circles, same as (a), and squares). The line shows T_C values calculated within the mean field model and for various assumptions about the hole spin susceptibility: the dashed line is for a 2D degenerate Fermi liquid, the dotted line takes into account the effect of non-zero temperature on a clean Fermi liquid, and the solid line is obtained assuming a gaussian broadening of the density-of-states. (c) Zero-field splitting of the PL line at 1.35 K, as a function of the hole density. The solid straight line is drawn through the data points; the dashed line is calculated within the mean-field model neglecting the effects of hole-hole correlation on the optical spectra. Both lines cross zero as shown by the dotted lines

the limited accuracy of our determination of the absolute value of the hole densities precludes any definitive conclusion. Nevertheless, it is worth noting that this linear dependence is markedly different from that found for (Zn,Mn)Te:N epilayers [16], in which a part of the Mn spins escape the ferromagnetic interaction at low carrier density, probably due to hole localization in the presence of acceptors in the DMS.

In conclusion, our results show how manipulating the density p of the two-dimensional hole liquid affects the ferromagnetic properties of magnetic quantum wells. In particular, in agreement with the theoretical expectations [7], the spontaneous magnetization is linear in p. While no dependence of T_C on p is expected for a fully degenerate ideal 2D hole liquid, we observe a dependence which is rather weak and well explained by disorder effects. Moreover, our findings show that both photon beam and electric field can isothermally drive the system between the ferromagnetic and paramagnetic phases, in a direction which can be selected by an appropriate design of the structure. This offers new tools for patterning magnetic nanostructures as well as for information writing and processing, beyond the heating effects of light exploited in the existing magneto-optical memories. Obviously, however, practical application of the tuning capabilities put forward here have to be preceded by a progress in the synthesis of functional room temperature ferromagnetic semiconductors. As far as II-VI compounds are concerned, according to theoretical suggestions [17, 18], structures containing ZnO, such as p-type (Zn,Mn)O/(Zn,Mg)O, appear to be a prospective material system.

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